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Thoughts about nano dosimetry.

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Abstract

The measurement of energy deposition in very small volumes has been called *nanodosimetry*. The concept has appeared in the scientific literature for some time. In this study it is shown that very little knowledge can be gained from nanodosimetry.

I. Introduction.

The observation of the energy loss, energy deposition and radiation effects of fast charged particles in *small volumes* is of interest in fields such as microdosimetry, radiation biology, materials science and high energy particle physics. I want to use the expression *signal* to describe the effect observed in a detector. In most measurements the signal is produced by the energy deposition rather than the energy loss. Here I want to consider volumes in which the particles make fewer than ten collisions on the average. Then the classical Landau¹ and Bethe-Bloch ideas² are not applicable. Concepts such as mean energy loss and absorbed dose are not suitable to relate energy loss and radiation effects in small volumes because energy loss or energy deposition spectra show a very large spread and consequently radiation effects will show large variations, too. As a corollary we note that it may not be necessary to obtain fine detail of the collision cross sections used in the simulation of energy loss.

Radiation effects in small volumes have been described for over thirty years in the symposia on microdosimetry, most recently in.³ The problems I want to consider here occur in the investigations for *very small volumes*, as for example that of a DNA structure.

Many effects encountered in radiation biology are discussed by Lea.⁴ This book should be studied in detail by all researchers in this field.

For detectors of high energy radiation attention must be paid to selecting a proper size of the detector. In superconducting wires with diameters of less than 1 μm , or in Ar gas of thickness less than 1 mm, no interactions will occur for some traversing particles, or the radiation signal is too small to be observed, and the particles will not be detected, even though the mean energy loss may exceed 300 eV.

II. Interactions of charged particles with matter.

These interactions are inherently stochastic in nature: collisions take place at random intervals and in each collision a random energy loss occurs. The probability of collisions in traversing a length x of material by particles with charge ze and speed $v = \beta c$ is given by the Poisson distribution ¹

$$P_n = \frac{m_c^n}{n!} e^{-m_c} \quad (1)$$

where n is the number of collisions *one* particle experiences, $m_c = x/\lambda$ is the average number of collisions particles suffer and λ (which depends on v) is the mean free path between collisions. The probability for an energy loss E is given by the differential collision cross section $\sigma(E)$

¹The dependence on v will not be indicated explicitly in the expressions for $m_c, \lambda, \sigma(E), f(\Delta; x)$ etc.

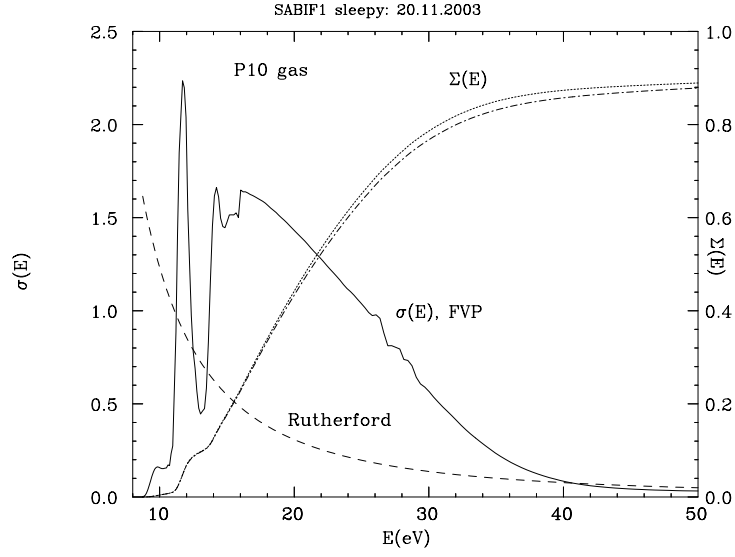


Figure 1: Differential collision cross section $\sigma(E)$ for single collisions of charged particles with speed $\beta\gamma = 3.6$ in P10-gas, as calculated with the Fermi-virtual-photon method (FVP)^{8,11}: solid line. The function extends to a value $E_{Max} \sim 12$ MeV. For present purposes only the part shown here is relevant. Except for the scale of the ordinate, the function depends very little on particle speed. The Rutherford cross section which is used for the Vavilov-Landau calculation is given by the dashed line. It is normalized to give the same stopping power as $\sigma(E)$. The integral function $\Sigma(E)$, Eq. (2) for $\beta\gamma = 3.6$, is shown by the dotted line, that for $\beta\gamma = 0.1$ by the dash-dotted line. Note the small difference for a large change in speed and that for only 12% of the collisions an energy loss exceeding 50 eV will happen, also see Fig. 3 in⁶ or Fig. 5 in.⁹

and its integral

$$\Sigma(E) = \int_0^E \sigma(E') dE' / \int_0^\infty \sigma(E') dE' \quad (2)$$

which is used for Monte Carlo simulations. Examples of $\sigma(E)$ and $\Sigma(E)$ for P10 gas are shown in Fig. 1. The moments of $\sigma(E)$ are

$$M_\nu = \int_0^\infty E^\nu \sigma(E) \cdot dE \quad \nu = 0, 1, 2... \quad (3)$$

Note that λ in Eq. (1) is equal to $1/M_0$. The “Bethe-Bloch equation”² is an evaluation of M_1 . For singly charged particles with $\beta\gamma = 3.5$, near minimum specific ionization, traversing P10 gas, the total collision cross section is $M_0 = 30$ collisions/cm and the stopping power is $M_1 = 2.56$ keV/cm. For Si, $M_0 = 4$ collisions/ μm , $M_1 = 0.4$ keV/ μm , and for liquid water $M_0 \sim 2.5$ collisions/ μm , $M_1 \sim 0.2$ keV/ μm .

P10 gas (90% Ar and 10% methane) is used in high energy particle ionization chambers. The structure of this spectrum is similar to that of ice,⁵ except for the spike of excitation to the first excited state at 12 eV. Spectra for Si can be found in,⁶⁻⁸ for water in.⁹

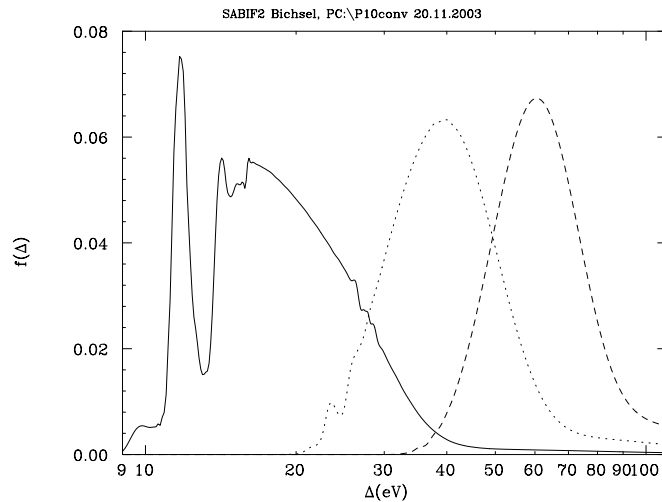


Figure 2: The convolution spectra $\sigma(E)^{*n}$ defined in Eq. (5) for P10-gas are given. Solid line: single collisions, dotted line: two collisions, dashed line: three collisions.

The collision cross section $\sigma(E)$ is different for each material, but the general features are similar: there is a pronounced peak between 12 and 25 eV which, for solids, is ascribed to a collective- or plasmon- excitation, there are further peaks at higher energies, usually broader and related to excitations of M-, N-, O- shells, and further well defined peaks related to excitations of inner-shell electrons.¹⁰ It is one of the purposes of this study to show that for the descriptions of radiation effects considered here the *detailed structure of the collision spectrum is not important*.

The *total* energy loss spectrum $f(\Delta)$ in an absorber of thickness x is derived in the next section.

III. Calculated energy loss spectra (“straggling functions”) $f(\Delta; x)$

Straggling functions can be calculated with computer-analytic methods^{12,7}

$$f(\Delta; v, x) = \sum_{n=0}^{\infty} \frac{m_c^n e^{-m_c}}{n!} \sigma(\Delta; v)^{*n} \quad (4)$$

where

$$\sigma(\Delta)^{*n} = \int_0^{\Delta} \sigma(E) \cdot \sigma^{*(n-1)}(\Delta - E) \cdot dE \quad (5)$$

is the n -fold convolution of the single collision spectrum $\sigma(E)$. The particle speed v is implicit as a parameter. The convolution spectra of Eq. (5) for P10 are given in Fig. 2. Note the large reduction of the 12 eV spike for $n = 2$, and its complete disappearance for $n = 3$. The spectra for Si are given in Fig. 3.

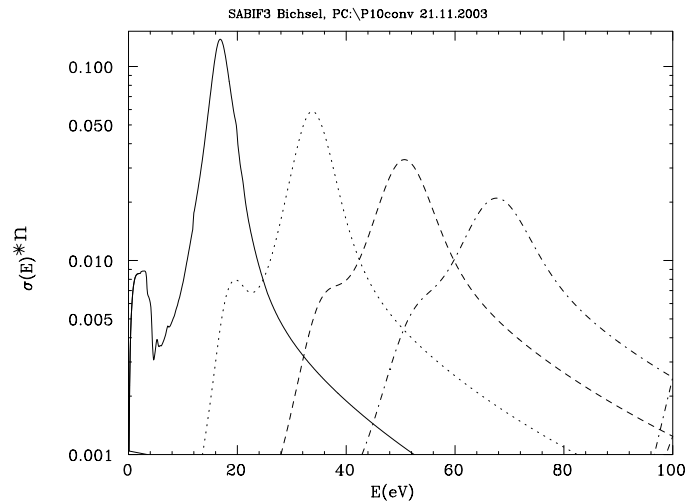


Figure 3: Same as Fig. 2 for solid Si, but $\sigma(E)^{*4}$ is also shown. The “plasmon peak” at 17 eV appears again at $17n$ eV, and its FWHM is proportional to n . The structure at ~ 2 eV also appears at $2 + 17(n - 1)$ eV, but diminishes with increasing n .

The energy loss spectrum $f(\Delta; v, x)$, Eq. (4), for particles with $\beta\gamma = 3.6$ passing through $x = 1$ mm in P10 is given by the solid line in Fig. 4, its integral, $F(\Delta) = \int_0^\Delta f(\Delta')d\Delta' / \int_0^\infty f(\Delta')d\Delta'$, by the dashed line. Here, $m_c = 3$, $P_0 = 0.05$, $P_1 = 0.15$, $P_2 = 0.22$, $P_3 = 0.22$, $P_4 = 0.17$, $P_5 = 0.10$, $P_6 = 0.05$. Thus, 5% of all particles will make no collisions at all, and the single collision spectrum is clearly seen, while most of the structure of $\sigma(E)$ has disappeared for multiple collisions.² The mean energy loss for the full spectrum is 260 eV, while for 60% of the collisions the energy deposition Δ is less than 100 eV.

Energy loss spectra for thin absorbers can be measured with electron microscopy or related methods.^{13,5}

IV. A simulated energy deposition spectrum $g(\Delta, x)$

In this section I want to demonstrate fundamental problems encountered in *measuring* radiation signals or effects. In order to show that these problems cannot be avoided even with a very good detector, the example of a “measurement” is made with a Monte Carlo simulation on a computer, using the following assumptions

- a) energy deposition and energy loss are the same
- b) the energy loss spectrum $F(\Delta)$ shown in Fig. 4 is binned into 0.1 eV intervals,
- c) values of the energy loss Δ are obtained with random numbers from $F(\Delta)$

²This is different for materials with a narrow plasmon peak, e.g. Al or Si.^{12,8}

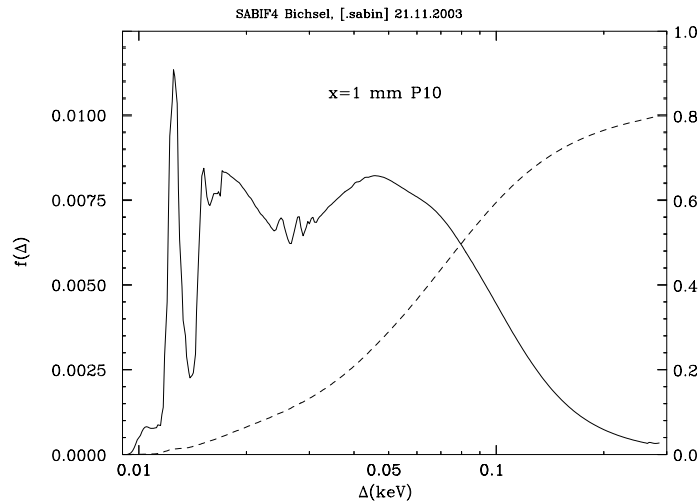


Figure 4: The energy loss spectrum for minimum ionizing particles passing through $x = 1$ mm of P10-gas is given by the solid line. No collisions will occur for 5% of the particles. Single collisions occur between 10 and 20 eV, and a small structure remains at 25 eV, but 2 and 3 collision spectra (as shown in Fig. 2) are merged at 50 eV. The integral spectrum $F(\Delta)$ shows that only 20% of the particles lose more than the mean energy loss $\langle \Delta \rangle = 250$ eV.

- d) the hypothetical detector inside the absorber produces *signals* s which are binned into 1 eV intervals, resulting in a distribution function $g(s)$ which can be represented by a histogram;
- e) the passage of 100,000 particles is simulated

Two examples, calculated with a different initial value for the random number generator are shown in Fig. 5 by crosses and circles. As we must expect, the two “measured” spectra differ. I have made calculations with 1,000,000 particles, resulting in a similar aspect, but with smaller variations. The important observation we can make is that *the detailed features of the energy loss spectrum are smeared out in the “observed” spectrum*. The most important reason for this is the random uncertainty due to the finite number of signals for each bin. A secondary reason is the loss of resolution due to the larger bins for the “measurement”.

For realistic detectors, this smearing will be more pronounced.

V. Realistic relation of energy loss to radiation effect.

In order to derive the radiation-signal or -effect from energy deposition spectra we must consider the nature of the detector. A fundamental problem for all detectors is the escape of secondary radiation, such as delta-rays or fluorescence photons from the volume under observation.^{14–16} It is neglected here.

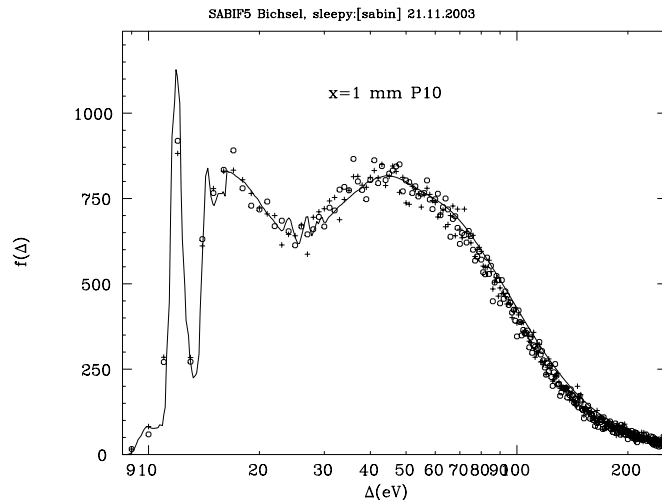


Figure 5: “Measured spectra” for a hypothetical detector described in the text are shown by circles and crosses. The energy loss spectrum of Fig. 4 is shown by the solid line. The “Measurement” is simulated with a Monte Carlo calculation. Instead of showing the histogram $g(s)$, the central point for each bar is given.

Let us consider two detectors schematically.

A. Ionization devices.

For both solid state and gaseous detectors, a minimum energy is needed to produce a signal, i.e. an ion-pair or an electron-hole pair. For the energy deposition spectrum in P10 (for which the energy needed to produce one ion pair is $w = 25 \text{ eV}^{17}$), postulated in Fig. 5, this means, schematically, that all energy losses between 12 and 25 eV are represented by *one* ion pair, energy losses between 25 and 50 eV are represented by *two* ion pairs etc., i.e. the spectrum of ion pairs will be represented by δ -functions at 22, 40, 65, 90 eV etc. and no other energy deposition effects can be measured. This “observable spectrum” is shown in Fig. 6. The exact process of the ionization by a given energy deposition is not relevant: while an energy deposition of 125 eV might produce as many as 10 ion-pairs (on the average 5), signals will always occur at integral numbers of ion-pairs; alternatively, a signal of 5 ion-pairs could be caused by any energy deposition between 70 to 140 eV. Clearly, any fine structure of the *energy loss* spectrum cannot be seen. ³

For a Si detector (where $w = 3.6 \text{ eV}$), the fact that the most probable energy loss is 17 eV means, schematically, that a single collision on the average produces 5 electron-hole pairs so

³In principle, M_0 or λ can be calculated precisely for a given $\sigma(E)$, thus will change even with small changes in $\sigma(E)$, but no differences in energy deposition will be discernible because of the random nature of their distribution.

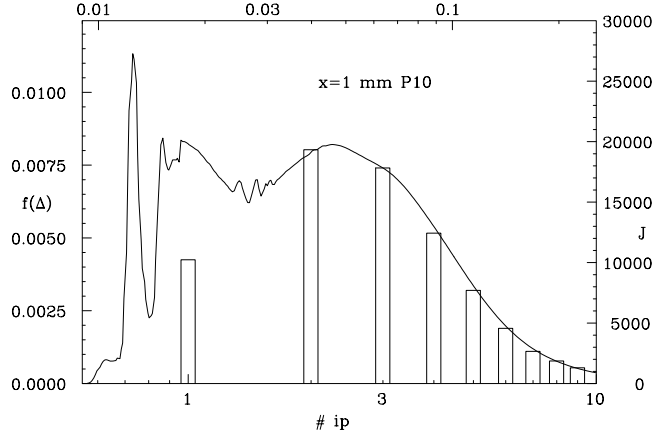


Figure 6: The “observable spectrum” for an ionization detector described in the text is shown by the bars representing the frequency (right hand scale) of signals from $\#ip$ ion pairs. The energy loss spectrum of Fig. 4 is shown by the solid line. The “observation” is derived from the Monte Carlo spectrum shown in Fig. 5. Clearly, very little about the energy loss spectrum could be derived from the observation. In a real world detector, several sources of “noise” would cause a spread in the bar give here, resulting in a Polya distribution for $n = 1$, approaching a Gaussian for large n .¹⁸

that the δ -functions will be located at multiples of 17 eV.¹⁶ The complication that the processing of each signal additionally is smeared by amplifier noise¹⁸ will disguise the discrete nature of the radiation effect.

B. DNA structures.

If DNA is considered as a detector, the situation is much more complex because the absorber is inherently highly inhomogeneous and the location of the collisions relative to the DNA will influence the probability for finding a break in a strand, as will the spatial extent of the collective excitations and their magnitude. These problems are considered in a study by Chatterjee and Holley.¹⁹ A second problem is caused by the particle fluence, the energy loss spectrum will have random numbers of losses in any given interval $[\Delta, \Delta + \delta\Delta]$ and in any given volume. While it appears that energy transfers of the order of 2 or 3 eV are sufficient to produce a single-strand break, the most probable energy losses in water⁹ and organic molecules²⁰ are the collective excitations at about 20 eV, enough to cause several DNA breaks in the neighborhood of the location of the collisions. Clearly, it will be difficult to correlate the collective excitations in water with any specific DNA break. Furthermore, radiation effects on DNA will also be

caused by the free radicals produced as a by-product of the energy depositions. Thus at the level of single collisions, the radiation effect will essentially be completely random.

VI. Conclusions.

The experimental measurement of radiation effects in physical detectors with very small volumes will not produce any meaningful results for the determination of biological effects. In particular the attempt to correlate physical measurements with biological effects does not make sense because the effect mechanisms are different. It allows only very coarse conclusions about the nature of the energy loss process. In particular very little can be derived about the structure of the differential collision cross section. Nano-dosimetry in the meaning of measuring spectra of radiation signals in small volumes is a futile exercise.

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